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16. ABSTRACT The paper gives results of pilot plant experiments to investigate the feasibility of replacing makeup water in limestone flue gas desulfurization (FGD) scrubbers with a simulated cooling tower blowdown. Tests were conducted while forcing the oxidation of the scrubber slurry to gypsum and maintaining Cl(-) concentrations at levels expected with the use of high-sulfur coals of moderate chloride content. Results show that all of the makeup water could be replaced with blowdown containing up to 690 ppm Na(+) when operating at SO2 make-per-pass below 8 mmol/L and a tightly closed scrubber loop. Material balances indicate that the uncontrolled discharge of soluble salts and trace elements from a power plant can be markedly rediced by application of water treatment units such as vapor-compression/evaporation as an integral part of the FGD scrubber. Such systems should be more effective in maximizing water reuse and should have lower energy requirements than the direct application of water treatment to blowdown streams.

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Desulfurization	Scrubbers		07A,07D	13I
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Combined Flue Gas Desulfurization and Water Treatment in Coal-Fired Power Plants

EPA-600/J-80-004
JOURNAL ARTICLE

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■ Pilot plant experiments were carried out to investigate the feasibility of replacing makeup water in limestone FGD scrubbers with a simulated cooling tower blowdown. Tests were conducted while forcing the oxidation of the scrubber slurry to gypsum and maintaining Cl^- concentrations at levels expected with the use of high sulfur coals of moderate chloride content. Results show that all of the makeup water could be replaced with blowdown containing up to 690 ppm Na^+ when operating at SO_2 make-per-pass below 8 mmol/L and a tightly closed scrubber loop. Material balances indicate that the uncontrolled discharge of soluble salts and trace elements from a power plant can be markedly reduced by application of water treatment units such as vapor-compression evaporation as an integral part of the FGD scrubber. Such systems should be more effective in maximizing water reuse and should have lower energy requirements than the direct application of water treatment to blowdown streams.

During the past 2 years, extensive testing of prototype flue gas desulfurization (FGD) scrubbers has been carried out in the forced oxidation mode at a power plant burning high-sulfur U.S. coals. The results (*1, 2*) demonstrate that complete oxidation of the waste sludge can be accomplished efficiently and reliably by air sparging the scrubbing slurry within the first loop of a two-loop venturi/spray tower system. It was shown that oxidation of the sulfite to calcium sulfate markedly improves the quality of the sludge: when the waste is separated by vacuum filtration, for example, its solids content is increased from 55% solids (as calcium sulfite) to 85% solids (as gypsum). Not only is the quality improved and the quantity of wet sludge reduced, but the filtration rate is also increased (*3*), requiring smaller equipment. Another important advantage of forced oxidation and sludge filtration is that it effectively eliminates sludge ponds from FGD systems, thereby avoiding potentially serious problems with the water balance due to excessive absorption of rainfall on large pond surfaces.

Pilot plant tests have shown (*4*) that forced oxidation can also be applied to single-loop limestone scrubbers, in which all of the scrubbing slurry is converted to gypsum within the scrubber effluent holding tank. In this case, additional benefits are foreseen: (a) improved control over gypsum supersaturation in the scrubbing liquor due to the large crystal seeding surface of the pure gypsum solids in the slurry; (b) simplification of the scrubbing chemistry by eliminating the formation of solid solutions of calcium sulfite/sulfate and minimizing the role of sulfite in the scrubbing reactions; and (c) elimination of oxidation as an otherwise uncontrolled variable which affects scrubber performance. The pilot plant tests also indicate that the aeration of the holding tank during oxidation can increase the pH and accelerate limestone dissolution (by stripping CO_2), effecting a slight improvement of SO_2 removal efficiency.

Whether one or two scrubbing loops are used, forced oxidation can be expected to improve the adaptability of a FGD unit for integration into the overall water management system in a power plant. This is because the makeup water can be replaced to some extent in such systems with cooling tower blowdown; the high density of pure gypsum seed crystals in the oxidized slurry can more rapidly dissipate any $CaSO_4 \cdot 2H_2O$ supersaturation induced by the extra sulfate from the cooling tower blowdown (which is primarily a solution of Na_2SO_4). One purpose of this paper is to examine the limits within which such a substitution might be made for the purpose of maximizing water reuse within the power plant as a whole.

The prospects for increasing water reuse, especially in FGD scrubber applications involving high chloride coals, are greatly improved by the successful demonstration of vapor-compression (VC) evaporation as a means of recovering pure water and concentrating the soluble salts in limestone scrubber effluents (*5*). Most coals contain chloride, which is discharged to the environment as HCl with boiler flue gas (when FGD is not used), or as soluble $CaCl_2$ in the waste sludge (when using limestone FGD scrubbers). Although some U.S. coals contain as much as 0.6% Cl, the average is about 0.1%, which corresponds to an uncontrolled discharge of 7 metric tons of $CaCl_2$ per day from a 500-MW power plant. In addition to its potential environmental impact, chloride can interfere with closed loop operation of FGD systems because it tends to promote corrosion, especially when present at concentrations above about 10 000 ppm in the scrubbing liquor. To prevent the accumulation of Cl^- to higher levels, it is common practice in Japan (*6*) to continuously purge a small stream of liquor from the scrubber as wastewater.

A second objective of this paper is to show that the application of VC water treatment—as an integral part of an FGD system—can moderate the Cl^- concentration to acceptable limits, even in the most tightly closed scrubber loops, and simultaneously control the discharge of soluble salts derived from both the cooling tower and coal combustion. The most efficient application of water treatment will thus be realized, with the greatest degree of water reuse and the least environmental contamination.

Experimental

A limestone scrubber of 7.5 m^3/min flue gas capacity (at a scrubber temperature of 50 °C) was set up in the single-loop configuration shown in Figure 1. The turbulent contacting absorber (TCA) was operated at a liquid/gas ratio of 10.1 L/m^3 and 3000 ppm SO_2 (dry basis) in the inlet flue gas, which also contained 6% oxygen and 8% H_2O. Chloride was fed as HCl gas with the flue gas to simulate the combustion of a coal of moderate Cl content, and maintain an average concentration of 13 000 ppm Cl^- in the scrubbing liquor. The oxidizer consisted of a 30-cm diameter tower containing slurry at a depth of 5.5 m, which was sparged with air at a stoichiometric ratio of 3.5 g-atoms of oxygen per g-mol of SO_2 absorbed. The air was injected at the bottom of the tower through pipes perforated with 6-mm diameter holes. A bleed stream of the oxidized slurry was processed by a rotary vacuum filter to remove the precipitated waste and return all filtrate to the scrubber in a fully closed loop mode. This pilot plant was operated 90 h continuously for each run.

The scrubbing slurry contained 8% solids (fly ash free), which was recirculated at a rate of 76 L/min. Ten complete liquid- and solid-phase analyses of the slurry were made during each run, in addition to flue gas analyses, to assess

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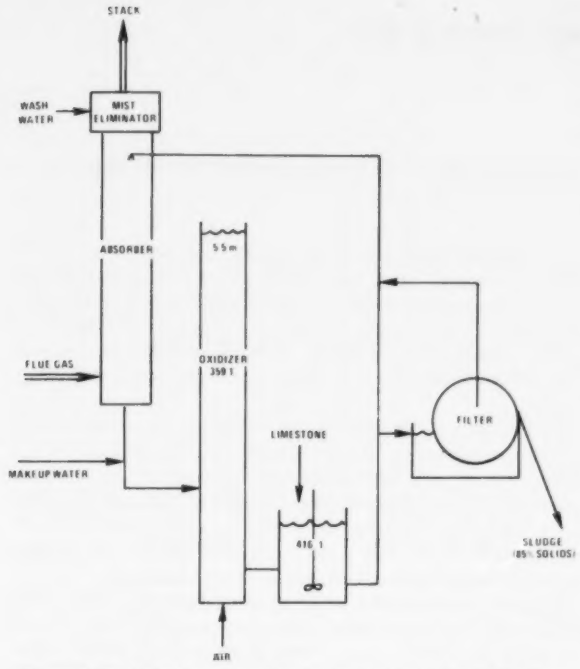

Figure 1. Pilot plant configuration for tests simulating water reuse

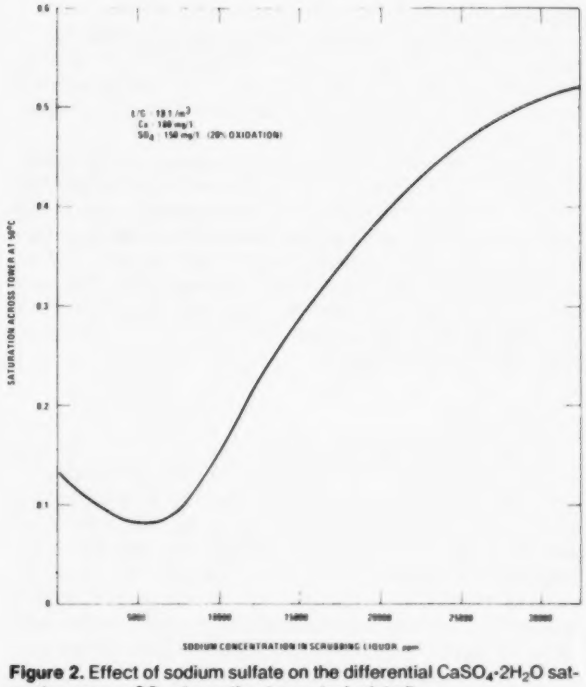

Figure 2. Effect of sodium sulfate on the differential $CaSO_4 \cdot 2H_2O$ saturation across SO_2 absorption tower (calculated)

overall performance in terms of limestone utilization, degree of oxidation, gypsum supersaturation, and SO_2 removal.

All of the fresh water normally fed to the scrubber (as makeup for evaporation losses and sludge moisture) was replaced with a simulated cooling tower blowdown consisting of Na_2SO_4 dissolved in water saturated with gypsum. This solution was fed to the scrubber effluent slurry prior to oxidation. Because adipic acid has been shown to improve limestone scrubber performance significantly without increasing the operating cost (7), supplementary tests were made with

Table I. TCA Scrubber Performance with and without Sodium Sulfate Addition[a]

	freshwater makeup	Na_2SO_4 solution makeup
SO_2 removal, %	86	91
limestone utilization, mol %	84	85
Cl^- concn in liquor, ppm	17 000	23 000
Na^+ concn in liquor, ppm	0	12 000
scrubber feed pH	6.0	6.6
scrubber effluent pH	5.0	5.6
oxidation, mol % SO_2 absorbed	99	99
slurry settling rate, cm/min	2.5	2.9
gypsum saturation ratio in scrubber feed liquor	1.0	1.11
absorber scale?	no	no

[a] Scrubber ΔP = 21 cm H_2O; inlet SO_2 = 3000 ppm.

this buffering additive; it was fed by dissolving in the sodium sulfate solution.

Results

The SO_2 removal efficiencies obtained while feeding the sodium sulfate were better than those obtained with fresh water makeup. As indicated by the comparison in Table I, the improvement amounted to 5% when operating at equivalent levels of limestone utilization. When compared at equal molar feed ratios of limestone/SO_2, the tests with Na_2SO_4 yielded about 8% higher limestone utilization and 2% higher SO_2 removal. The comparisons were not made in sufficient number to quantify the statistical significance of these differences. All tests showed consistently higher scrubber feed pHs when the makeup water was replaced with sodium sulfate solution.

Complete chemical analysis of the scrubber feed liquor during eight consecutive runs showed an average saturation ratio of 1.13 for gypsum, as determined by the Bechtel-modified Radian equilibrium program (8). Operating at a sodium level of 15 000 ppm in the scrubbing liquor, gypsum scale was consistently observed on the bottom TCA grid when the SO_2 make-per-pass exceeded 8 mmol/L. The system could be operated scale-free at this sodium level by reducing the SO_2 make-per-pass below 8 mmol/L, e.g., by reducing the inlet SO_2 concentration in the flue gas to 2500 ppm.

At a sodium level of 8000 ppm in the scrubbing liquor, the system operated scale-free when the inlet SO_2 concentration was 3000 ppm in the flue gas. When adipic acid was added to the scrubbing liquor at a level of 630 ppm, performance comparable to that of Table I (88% SO_2 removal at 85% limestone utilization) required only a 14-cm water pressure drop in the TCA—30% less than the pressure drop required without the additive. No adverse effects on scaling potential or oxidation efficiency were observed with the use of adipic acid.

An overall oxidation rate of 1×10^{-3} g-mol/L-min was maintained in the oxidizer at an average pH of 6.5 when operating with 13 000 ppm Cl^- and 15 000 ppm Na^+ in the scrubbing liquor. The oxidized sludge was excellent quality, consistently filtering to 85% solids and settling at a rate of 2.9 cm/min at 50 °C. It contained 2 mg of sodium per g of dry solid when washed with acetone. About 0.3 mg of this sodium was nonleachable by water washing.

Discussion of Results

The apparent effect of sodium sulfate on SO_2 removal can be explained by laboratory studies (9) that show the rate of SO_2 absorption is increased by the presence of dissolved salts: ion diffusivities and the equilibrium constant are favorably affected, resulting in about 20% greater SO_2 absorption rate

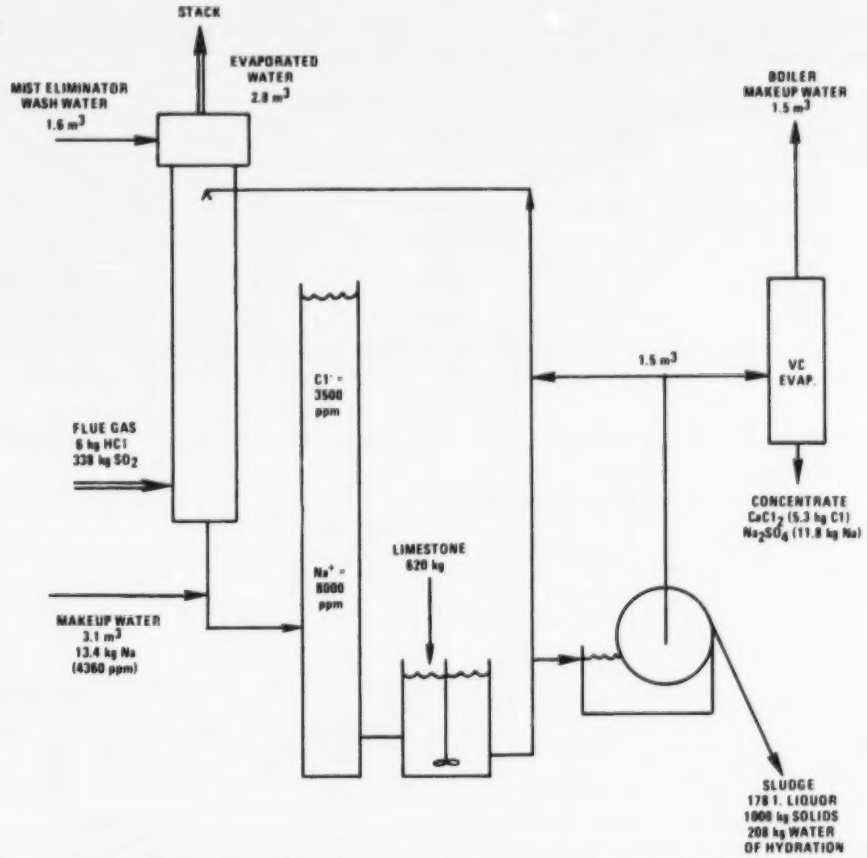
STACK
EVAPORATED WATER 2.8 m³
MIST ELIMINATOR WASH WATER 1.6 m³
BOILER MAKEUP WATER 1.5 m³
1.5 m³
VC EVAP.
Cl⁻ = 3500 ppm
FLUE GAS 6 kg HCl 338 kg SO₂
CONCENTRATE CaCl₂ (5.3 kg Cl) Na₂SO₄ (11.8 kg Na)
LIMESTONE 620 kg
Na⁺ = 8000 ppm
MAKEUP WATER 3.1 m³ 13.4 kg Na (4360 ppm)
SLUDGE 178 l. LIQUOR 1000 kg SOLIDS 208 kg WATER OF HYDRATION

Figure 3. Water balance for FGD system applied to boiler burning coal containing 4% S and 0.1% Cl; VC evaporator extracting 89% of salts from scrubbing liquor. All makeup water replaced with cooling tower blowdown containing 4360 ppm Na^+

into 0.4 M NaCl than into water. The salt concentrations used here were within the range shown to be effective.

The observed effect of sodium sulfate on scaling potential is explained by the relationship shown in Figure 2 where the differential $CaSO_4 \cdot 2H_2O$ saturation across the absorber, as computed with the Bechtel/Radian program, is plotted as a function of sodium concentration. It assumes that 20% of the SO_2 is oxidized in the TCA and the Ca^{2+} concentration rises 180 ppm due to the limestone dissolution in the absorber—corresponding to a make-per-pass of 9 mmol/L and 50% dissolution in the absorber. The calculations and test results agree that scaling can be expected at sodium levels approaching 15 000 ppm; above that level the critical supersaturation ratio (*10*) of 1.35 will be exceeded at the bottom of the absorber, and scaling can occur.

The results indicate that scrubbers applied to boilers using low sulfur coal—which operate at SO_2 make-per-pass well below 8 mmol/L—should be able to accommodate sufficient sodium in the makeup water to yield a steady-state level of 15 000 ppm in the scrubbing liquor. A material balance on such a system operating in the mode of Figure 1 (in which the liquor purged as moisture in the filter cake amounts to 178 L per metric ton of dry sludge produced) shows that this situation will occur when the sodium feed is 2.6 kg/ton of dry sludge produced, or the makeup water is totally replaced by a blowdown containing 690 ppm Na. This sodium level is within the limits of most blowdown compositions. If the scrubber were applied to a boiler using high-sulfur coal, it could accept up to 370 ppm Na in the blowdown without exceeding the 8000-ppm limit that these tests indicate is safe. In either case, a chloride balance shows that the maximum allowable chloride content of the coal will be 0.03%, if a steady-state Cl^- level of 10 000 ppm is not to be exceeded in the scrubber.

Salt Extraction. Water treatment units are currently applied directly to cooling tower and boiler blowdown streams in some U.S. power plants to extract soluble salts and recover pure water. Typical compositions of these streams are described by Anderson (*11*). Vapor-compression evaporation units capable of processing 680 L/min at concentration factors up to 75 are operational (*11, 12*). The application of such units to a FGD system, rather than directly to the blowdown, would permit the simultaneous extraction of soluble salts that accumulate in the scrubber loop (e.g., $CaCl_2$) as well as those from the cooling tower (Na_2SO_4). Such an arrangement, illustrated in Figure 3, could markedly reduce the total amount of soluble salts released to the environment. It would also effect suitable control over the concentration of salts within the scrubbing loop to prevent gypsum scaling and avoid chloride corrosion, even when burning coals of the highest Cl content. Most importantly, it would isolate the soluble salts, including trace elements, so that they are not disposed of with the sludge. Waste treatment technology can be applied more effectively and cheaply to a small concentrated stream than to the entire mass of sludge produced by the scrubber.

To assess the effects of incorporating water treatment into the FGD system, material balances were calculated for several assumed modes of operation. Since vapor-compression evaporation has been demonstrated as a feasible method of concentrating waste liquor from a FGD scrubber (*5*), it is assumed that this method is applied to a slipstream of clarified liquor for salt extraction. The material balance for this system is:

Table II. Effect of Water Treatment on the Chloride Concentration in the Scrubber Liquor and Total Salt Extracted from FGD Systems, Calculated for Three Different Methods of Sludge Removal[a]

evaporator throughput, L/ton of dry FGD sludge	40% [b] solids in sludge		55% [c] solids in sludge		85% [d] solids in sludge	
	chloride, ppm	salt recovery, %	chloride, ppm	salt recovery, %	chloride, ppm	salt recovery, %
0	5000	0	9200	0	33 000	0
250	4100	18	6600	28	14 000	58
500	3800	25	5700	38	8 600	74
750	3300	33	4800	48	6 300	81
1500	2500	50	3200	65	3 500	89
2500	1900	63	2300	76	2 200	93

[a] Basis: 0.1% Cl, 4% S in coal. [b] Settler/clarifier, no forced oxidation. [c] Vacuum filter, no forced oxidation. [d] Vacuum filter, with forced oxidation.

Na fed with blowdown + Cl absorbed from flue gas
= salt in liquor contained in sludge
+ salt in evaporated slipstream

where the total salt content of each stream is the product of the salt concentration and the total stream flow. Table II summarizes the results of material balances for three different methods of sludge removal: (a) settling of sulfite slurry, (b) vacuum filtration of sulfite slurry, and (c) vacuum filtration of gypsum. All balances are based on 1 metric ton of dry oxidized solids produced by FGD, or the equivalent 750 kg of unoxidized solids.

Several conclusions can be drawn from the comparisons in Table II. First, it is clear that the soluble salts can be extracted more efficiently from a system employing forced oxidation than without it. For example, by treating 1.5 m^3 of clarified liquor per ton of dry FGD sludge produced, about 50% of the soluble salts could be extracted from a scrubber which discharges unoxidized sludge containing 40% solids. If the same amount of liquor is processed while forcing oxidation (and discharging sludge containing 85% solids), 89% of the soluble salts are extracted. The same percentages of reduction indicated in Table II for soluble salts also apply to the heavy metals and trace elements scrubbed from the flue gas by the FGD system. These potentially harmful products of coal combustion would be isolated from the FGD sludge and concentrated in the same stream with the soluble salts.

Since the pilot plant tests and Figure 2 both indicate that a sodium level of 8000 ppm is a realistic operating condition for most FGD systems using forced oxidation—regardless of the type of coal burned in the power plant—the material balances were extended within that constraint assuming that VC is applied to a slipstream of the filtrate liquor to extract 89% of the salts. Figure 3 shows the water balance for such a system, in which all of the makeup water is replaced with cooling tower blowdown. It is assumed in this water balance that the limestone feed slurry is made up with clarified liquor, and that the water used for washing the mist eliminator is not replaced with blowdown since high quality water is required for that purpose. In accordance with the washing procedure established for scale-free operation of mist eliminators (*13*), 1.6 m^3 of fresh water is used per ton of dry, oxidized FGD sludge produced. This water is applied intermittently to the top and bottom of the mist eliminator at an average rate of 4 L/min per m^2 of cross-sectional area.

A combined FGD/water-treatment system operating as shown in Figure 3 can accept blowdown containing as much as 4360 ppm Na^+ (or 9100 ppm sulfate) without exceeding the 8000-ppm Na^+ limit in the scrubbing liquor. Few, if any, blowdown streams would approach this concentration. The chloride concentration in the scrubbing liquor would be only 3500 ppm when the system is applied to a boiler burning coal containing 0.1% chloride. Figure 4 gives the chloride concentrations expected in the scrubbing liquor, as a function of VC throughput, when coals of other Cl contents are burned. It assumes that the makeup water (or blowdown) contains no chloride and that no chloride is coprecipitated in the gypsum. Laboratory analyses of well-washed pilot plant sludge support the latter assumption (Cl < 0.3 mg per g of dry gypsum).

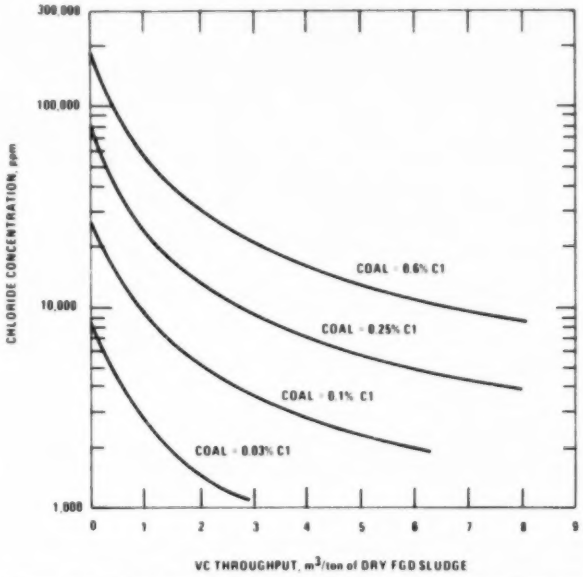

Figure 4. Chloride concentration in FGD scrubber liquor as a function of VC evaporator throughput and the chloride content of coal burned in the power plant; sludge = 85% solids

The system shown in Figure 3 requires a VC throughput of 1.5 m^3 per ton of dry FGD sludge produced. In accordance with Table II and Figure 4, higher VC throughputs would further increase the isolation of soluble salts in a concentrated bleed stream—with additional energy penalty. Alternate methods of increasing the extraction efficiency would be: (a) to apply compression rollers to the vacuum filter to raise the solids content of the cake to 92% (*14*), and/or (b) replace only 90% of the makeup water with blowdown using the remainder for filter cake washing. In the latter case, VC feed would be drawn from one unwashed filter module.

When applied to a 760-MW power plant, which produces about 54 tons of dry gypsum per hour, the combined system shown in Figure 3 would require two standard-size VC units with a total throughput of 1.36 m^3/min. The electric power consumed by these units, when designed to operate at a concentration factor of 140, is in the range of 2.6–3.2 kWh/m^3 (5)

or about 2360 kW for the power plant. This power requirement amounts to 0.31% of the total electric power produced by the plant, which compares to 1.3–4.0% consumption of total output by a well-designed FGD scrubber (the precise amount depends mainly on the type and degree of flue gas reheat used).

It is current practice to apply VC directly to cooling tower blowdown streams as a means of accomplishing water reuse in power plants. A comparison of that approach with the method discussed here shows that the integration of VC into the FGD system while using blowdown to replace FGD makeup water is a more efficient procedure: direct VC treatment of the blowdown stream shown in Figure 3 would require evaporation of 3.1 m^3 instead of the 1.5 m^3 required by the combined FGD/water-treatment system. This improvement results primarily from the fact that the FGD scrubber serves as an evaporator to preconcentrate the blowdown prior to VC treatment. Considering the power plant as a whole, water recovery is also increased nearly 50% (1.5 m^3 of VC condensate plus 3.1 m^3 of fresh water replaced by blowdown), while producing sufficient high-quality condensate to meet the needs for boiler makeup water.

In conclusion, this analysis shows that the application of water treatment to a FGD system can be expected to improve the efficiency of water reuse in power plants of the western U.S. and, in eastern applications, can facilitate the combustion of high-sulfur, high-chloride coals in an environmentally acceptable manner. In either case, such systems should enhance the prospects for controlling the discharge of potentially harmful soluble salts, trace elements, and heavy metals from coal combustion by isolating them in a manner that will permit more effective and permanent disposal methods to be applied.

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